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(54) SIZING AGENT COMPOSITION AND METHOD OF USING THE SAME.

(57) A sizing agent composition for paper comprising a mixture of (a) 70 to 99.9 wt % of substituted alkyl-succinic anhydride, substituted alkenylsuccinic anhydride or a mixture thereof and (b) 0.1 to 30 wt % of polyoxyethylene alkyl ether phosphate, polyoxyethylene alkylaryl ether phosphate or a mixture thereof, and a process for making paper by dispersing the composition in an aqueous solution of a water-soluble high-molecular compound, adding the dispersion to a pulp slurry, and making paper from the slurry. The composition has an excellent storage stability and self-emulsifiability in water, and exhibits a good sizing effect on paper.

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DESCRIPTION

TITLE MODIFIED

TITLE OF THE INVENTION:

see front page

COMPOSITION FOR SIZING AGENT AND PROCESS FOR USING
THE SAME COMPOSITION

TECHNICAL FIELD:

The present invention relates to a composition for a sizing agent consisting of a mixture of a substituted alkyl succinic anhydride or a substituted alkenyl succinic anhydride or a mixture of these succinic anhydrides, phosphates of polyoxyethylene alkyl esters or phosphates of polyoxyethylene alkyl aryl ethers or a mixture of these phosphates of esters and a process for using the composition for the sizing agent by dispersing the composition in aqueous solution of aqueous soluble polymer compound, adding the obtained aqueous dispersant in a pulp slurry of paper making raw material, and making paper and, more particularly, to a composition for sizing paper having excellent storage effect and preferable underwater self emulsion, and a process for making paper having excellent sizing effect by using the composition.

BACKGROUND ART:

There is known a sizing agent and particularly a

paper making sizing agent such as a mixture homogenized by mechanically agitating an aqueous solution of cationic starch and a substituted succinic anhydride in an aqueous dispersion to be mixed with a pulp slurry, a dispersion of mixture of premixing a substituted succinic anhydride and nonionic surfactant of certain type in water (in Japanese Patent Publication No. 36044/1978), or an aqueous dispersion of a substituted succinic anhydride and underwater oil type surfactant dispersed in a substituted succinic anhydride having diameters of ultrafine particles dispersed in aqueous solution of amphoteric acrylamide polymer (in Japanese Patent Laid-open No. 45,731/1983). A large scale of apparatus is required in case of producing the dispersion by using cationic starch of these aqueous dispersions, and yet it is difficult to sufficiently finely pulverizing the dispersion particles in the obtained aqueous dispersion. Therefore, the sizing agent has a drawback that the sizing effect of the dispersion is deteriorated, and the aqueous dispersions of the latter two having wrong storage stability of the composition before the aqueous dispersion of the latter two is dispersed in the aqueous dispersion, and as time is elapsed, the under water dispersant is erased, and the sizing performance is largely decreased, or not

performed as well. Consequently, the mixture composition must be adjusted immediately using as the sizing agent, and there is a problem in a practical use in an industrial scale.

In order to solve the above-described problems, the present inventors have proposed an aqueous dispersion containing a surfactant having a substituted succinic anhydride and a half ester residue of the substituted succinic acid (in Japanese Patent Application No. 154984/1982), but in the aqueous dispersion, when a substituted alkenyl succinic anhydride of an additive reaction product of a straight chain internal olefin and maleic anhydride is used as the substituted succinic anhydride, sufficient underwater dispersing effect cannot be obtained unless the above-described surfactant is mixed in large quantity, and since the large quantity of the surfactant substance becomes disturbing the sizing effect. Therefore, it is discriminated that excellent sizing effect cannot be obtained.

Accordingly, an object of the present invention is to provide a composition for a sizing agent which exhibits excellent storage stability and underwater self-emulsion and hence excellent sizing effect by consisting of (a) 70-99.9 wt.% of a substituted alkyl succinic anhydride or a substituted alkenyl succinic

anhydride or a mixture of these succinic anhydrides, (b) 0.1-30 wt.% of phosphates of polyoxyethylene alkenyl ether esters or phosphates of polyoxyethylene alkyl ether esters or a mixture of these phosphate esters, and a process for effectively making paper having excellent sizing effect by using the composition for the sizing agent, i.e., a process for using the composition for a sizing agent.

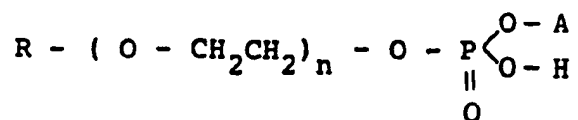
DISCLOSURE OF INVENTION:

A composition for a sizing agent of the present invention consists of (a) 70-99.9 wt.% of a substituted alkyl succinic anhydride or a substituted alkenyl succinic anhydride or a mixture of these succinic anhydrides, and (b) 0.1-30 wt.% of phosphates of polyoxyethylene alkyl ether esters or phosphates of polyoxyethylene alkyl aryl ether esters or a mixture of these phosphate esters.

A substituted alkyl succinic anhydride or a substituted alkenyl succinic anhydride in the composition for a sizing agent may employ any of compounds heretofore known per se as a sizing agent, and concretely employ a substituted succinic anhydride having 8 or more carbons and preferably 12-36 alkyl group or alkenyl group. This substituted succinic anhydride can be readily produced generally by utilizing alpha-olefin,

inner olefin or the additive reaction of an alpha-olefin, inner olefin or their mixture having carbon atoms of corresponding number or their mixture, and utilizing the additive reaction of these mixtures with maleic anhydride. Particularly, when the substituted succinic anhydride is, for example, octadecene-9, tetradecene-7, hexadecene-7, eicosane-11 or their mixtures, straight chain inner olefin mixture in which double bond produced by dehydrated reaction of a straight chain paraffin is substantially uniformly distributed at the positions except alpha-position, additive reaction product of inner olefin and maleic anhydride of straight chain inner olefin mixture having 70 % or more of total amount of inner olefin disposed at 2-, 3- and 4-position of double bond produced by anisotropic reaction in the presence of a catalyst with straight alpha-olefin, i.e., substituted alkenyl succinic anhydride having succinic anhydride group in a substituted group, the sizing effect effected by the composition for the sizing agent of the present invention becomes further excellent.

Other phosphates of polyoxyethylene alkyl ether esters or phosphates of polyoxyethylene alkyl aryl ether ester are preferably compounds represented by the following general formula:



where R is an alkyl or alkyl aryl group having 8 or more carbon atoms, A represents H, or

$R' - (O - CH_2CH_2)_m -$ (R' is alkyl or alkyl aryl group, m is 1 or more integer number), and n is 1 or more integer number. Particularly, when the R of the above general formula is alkyl or alkyl aryl group having 10 - 20 carbon atoms and the value of n is 5 or more, the underwater dispersion particles of the produced substituted succinic anhydride becomes extremely fine, and it is most preferable that the sizing effect executed by the composition of the present invention becomes very excellent.

In the mixture of (b) the phosphates of esters in the same manner as (a) the substituted succinic anhydride, the mixture of (a) 70 - 90 wt.% of substituted succinic anhydride and (b) 10 - 30 wt.% of the phosphates of esters produces water dispersion containing substituted succinic anhydride of sufficiently fine particle size by merely mixing the mixture, thereby providing so-called self-emulsion type paper making sizing agent. Further, the mixture of (a) 90 - 99.9 wt.% of substituted succinic anhydride and (b)

0.1 - 10 wt.% of the phosphates of esters disperses in aqueous solution of aqueous polymer compound the mixture, thereby producing aqueous dispersion of substituted succinic anhydride of fine particle size sufficiently stable by mixing means of the degree for extremely slowing agitating. Therefore, when the mixture of (a) the substituted succinic anhydride and (b) the phosphates of esters is used as the paper making sizing agent, it is preferable to use aqueous dispersion utilizing aqueous solution of aqueous soluble polymer compound due to the relationship of reducing the mixture amount of (b) the phosphates of esters so as to reduced the sizing effect disturbing action caused by (b) the phosphates of esters as small as possible.

According to the present invention a process for making paper of a process for using a composition for a sizing agent comprises adding aqueous dispersion that mixture of (a) 70 - 99.9 wt.% of substituted alkyl succinic anhydride or substituted alkenyl succinic anhydride or their mixture of these succinic anhydrides and (b) 0.1 - 30 wt.% phosphates of polyoxyethylene alkyl ether esters or phosphates of polyoxyethylene alkyl aryl ether esters or their mixture of these phosphates of esters in aqueous solution of aqueous soluble polymer compound to pulp slurry of paper making

material, making the paper in accordance with an ordinary method to provide paper.

In the aforementioned paper sizing process, the aqueous soluble polymer compound contained in the aqueous dispersion may employ not only cationic starch, ampholytic acrylic amide polymer heretofore known per se, but also other aqueous soluble starch derivatives, aqueous soluble cellulose derivatives, various acrylic amide polymers, polyvinylalcohol, polyethylene imine and its derivatives, aqueous soluble polyamide, aqueous soluble acrylic resin, aqueous soluble polyester, aqueous soluble maleic resin, and various aqueous soluble vinyl polymers. The aforementioned aqueous soluble polymer compounds contained in the aqueous soluble dispersion is preferably in a range of 0.1 - 5 wt. parts and preferably 0.5 - 2 wt. parts to 1 wt. parts of substituted succinic anhydride in the aqueous soluble dispersion.

In the process for making paper of process for using a composition for a sizing agent of the present invention, various medicines known in general in the sizing step of adding aqueous dispersion of the sizing agent into a pulp slurry such as paper strength intensifier, filtrate accelerator, filler yield agent, fixing agent or other sizing agent may be arbitrarily

used. As another process, the aqueous dispersion of the produced substituted succinic acid is coated on wet paper or semidried paper by suitably means known per se after making or sheet making, thereby enabling surface sizing.

The composition for a sizing agent of the present invention constructed as described above consists of a mixture of specific surfactant and substituted succinic anhydride. Since the surfactant has excellent solubility to the substituted succinic anhydride, excellent storage stability is provided irrespective of the type of the substituted succinic anhydride, stable underwater self-emulsion is provided, aqueous dispersion of substituted succinic anhydride having extremely fine particle size is provided in the presence of small amount of surfactant, and excellent operation is provided as the composition for the paper making sizing agent of various types.

Since a process for making paper of a method for using the composition for the sizing agent of the present invention adds the aqueous dispersion of substituted succinic anhydride having the above-described characteristics as sizing agent in paper making pulp slurry, it can eliminate the complexity of preparing the composition immediately before using

without large scale of emulsifying apparatus to produce aqueous dispersion. Therefore, it is effective to make paper having excellent sizing effect in an industrial scale.

Hereinafter, the present invention will be described further in detail by the preferred examples thereof as compared with reference examples of making paper utilizing the composition for the sizing agent and the aqueous dispersion of the composition of the present invention as the sizing agent, and the properties of the aqueous dispersion of the composition and the sizing effect of the paper will be moreover described.

Example 1, Comparison example 1

Substituted alkenyl succinic anhydride and surfactant listed in predetermined columns of Table 1 are utilized, both are agitated and mixed under slow heating condition at ambient temperature or 40°C or lower, and compositions (a), (b), (c), (d) for sizing agent of the present invention and compositions (1), (2) for comparison are prepared.

The substituted alkenyl succinic anhydride A is maleic anhydride reacted with inner olefin having 15-18 carbon atoms, and the substituted alkenyl succinic anhydride B is maleic anhydride reacted with oligomer mainly containing pentamer of propylene.

Table 1

| Composition | Substituted alkenyl succinic anhydride | Surfactant |
|-------------|---|---|
| (a) | A 80 wt. % | Phosphates of polyoxy- ethylene nonyl phenol ether ester (mono-, di- mixture) (polyoxyethylene: n=9) 20 wt. % |
| (b) | B 85 wt. % | ditto 15 wt. % |
| (c) | A 85 wt. % | Phosphates of polyoxy- ethylene nonyl phenol ether ester (mono-, di- mixture) (polyoxyethylene: n=21.5) 15 wt. % |
| (d) | B 90 wt. % | ditto 10 wt. % |
| (1) | A 80 wt. % | Polyoxyethylene nonyl phenol ether (polyoxy- ethylene: n=15) 20 wt. % |

| | | |
|-----|----------|---|
| | B | Sulfates of polyoxyethylene olenyl ether ester ammonium (polyoxyethylene: n=18) |
| (2) | 85 wt. % | 15 wt. % |

Experiment 1

1 wt. parts of compositions (a) to (d) and (1), (2) provided in the example and comparison example are mixed with 99 wt. parts of water, agitated, dispersed to prepare aqueous dispersions of different types.

The prepared aqueous dispersions are added to 2 wt. % pulp slurry containing 20 wt. % of pulp (L-BKP, c.s.F. 400cc) of heavy calcium carbonate filler (Escalon #800 made of Sankyo Powder Co.) so that the composition in the aqueous dispersion contains 0.3 wt. % of pulp, then cationic starch (Neo-Posiparin) made of Matsutani Chemical Industry Co.) of fixing agent is added to 0.5 wt. % of pulp, paper corresponding 80 g/m² is prepared in accordance with an ordinary process, dried in a rotary dryer of 110°C for 120 sec. to prepare paper.

The obtained paper is measured for sizing degree after moistened in accordance with JIS, and listed in Table 2.

Table 2 shows the compositions which are allowed

to stand for ten days, and the results of similar test to the previous examples are listed together with emulsion mean particle size (micron) of aqueous dispersions.

Table 2

| Composition | Prepared composition | | Composition after 10 days | |
|-------------|------------------------|---|---------------------------|---|
| | Stockigt sizing (sec.) | Mean particle size (micron) of emulsion | Stockigt sizing (sec.) | Mean particle size (micron) of emulsion |
| (a) | 38 | <1 | 36 | <1 |
| (b) | 20 | approx. 1 | 18 | approx. 1 |
| (c) | 46 | <1 | 40 | <1 |
| (d) | 23 | approx. 1 | 19 | approx. 1 |
| (1) | 30 | approx. 1 | 5 | 5-6 |
| (2) | 11 | approx. 1 | 0 | 5-6 |

Example 2, Comparison example 2

Substituted alkenyl succinic anhydride and surfactant listed in predetermined columns of Table 3 are utilized, both are agitated and mixed under slow heating condition at ambient temperature or 40°C or lower, and compositions (e), (f), (g), (h) for sizing agent of the present invention and compositions (3), (4) for comparison are prepared.

The substituted alkenyl succinic anhydride A is

maleic anhydride reacted with inner olefin having 15-18 carbon atoms, and the substituted alkenyl succinic anhydride B is maleic anhydride reacted with oligomer mainly containing pentamer of propylene.

Table 3

| Composition | Substituted alkenyl succinic anhydride | Surfactant |
|-------------|--|---|
| (e) | A 95 wt.% | Phosphates of polyoxy-ethylene nonyl phenol ether ester (mono-, di-mixture) (polyoxyethylene: n=9 5 wt.%) |
| (f) | B 96 wt.% | ditto 4 wt.% |
| (g) | A 95 wt.% | Phosphates of polyoxy-ethylene tridecyl ether ester (mono-, di-mixture) (polyoxyethylene: n=9 5 wt.%) |
| (h) | A 97 wt.% | Phosphates of polyoxy-ethylene nonyl phenol ether ester (mono-, di-mixture) (polyoxyethylene: n=21.5) 3 wt. % |

| | | |
|-----|-------------------|---|
| (3) | A 95 wt. % | Polyethylene nonyl phenol ether (polyoxy- ethylene: n=15) 5 wt. % |
| (4) | A 96 wt. % | Sulfates of polyoxyeth- ylene olenyl ether ester ammonium (polyoxyethy- lene: n=18) 4 wt. % |

Example 3, Comparison example 3

Compositions (e) to (h) and (3), (4) prepared in the example 2 and comparison example 2 are filled in 5 wt. % of aqueous solution of aqueous soluble polymer compound listed in predetermined columns of Table 4, agitated at 50V for 2 min. in a homomixer (HV-M type made of Tokushu Kika Kogyo Co), thereby preparing aqueous dispersions.

The prepared aqueous dispersions are added to 2 wt. % pulp slurry containing 20 wt. % of pulp (L-BKP, c.s.F. 400cc) of heavy calcium carbonate filler (Escalon #800 made of Sankyo Powder Co.) so that the composition of substituted alkenyl succinic anhydride and surfactant in the emulsion contains 0.5 wt. % of pulp, then cationic starch (Neo-Posiparin made of Matsutani Chemical Industry Co.) of fixing agent is added to 0.5 wt. % of pulp, paper corresponding 80 g/m² is prepared in accordance with

an ordinary process, dried in a rotary dryer of 110°C for 120 sec. to prepare paper.

The obtained paper is measured for sizing degree after moistened in accordance with JIS, and listed in Table 4.

Table 4 shows the compositions which are allowed to stand for ten days, and the results of similar test to the previous examples are listed together with emulsion mean particle size (micron) of aqueous dispersions.

Table 4

| Composition | Aqueous polymer | Part of solution to part of composition (part) | Prepared composition | | Composition after 10 days | | |
|-------------|-----------------|--|------------------------|---|---------------------------|---|-----------|
| | | | Stockigt sizing (sec.) | Mean particle size of emulsion (micron) | Stockigt sizing (sec.) | Mean particle size of emulsion (micron) | |
| (1) | (e) | Cationto Starch | 40 | 37 | 1 | 31 | 1 |
| (2) | (e) | 90 mol% of acrylamide, 10 mol% of dimethylamino-ethylmethacrylate copolymer | 10 | 24 | 1 | 22 | 1 |
| (3) | (f) | ditto to (1) | 40 | 20 | approx. 1 | 19 | approx. 1 |
| (4) | (g) | ditto to (1) | 40 | 33 | 1 | 30 | 1 |
| (5) | (h) | 95 mol% of acrylamide, 5 mol% of dimethylamino-ethylmethacrylate benzyl chloride | 10 | 28 | approx. 1 | 27 | approx. 1 |
| (6) | (3) | ditto to (1) | 40 | 30 | 1 | 19 | 3-4 |
| (7) | (4) | ditto to (1) | 40 | 32 | approx. 1 | 17 | 4-5 |

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INDUSTRIAL APPLICABILITY

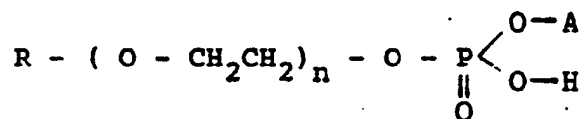
As described above, the composition for the sizing agent according to the present invention and the process for using the composition provide excellent storage stability, underwater self-emulsion, and accordingly useful as the composition for the sizing agent exhibiting the sizing effect, and advantageous as the process for using the composition for making paper having excellent sizing effect by using the composition.

CLAIMS:

1. A composition for a sizing agent comprising (a) 70-99.9 wt.% of a substituted alkyl succinic anhydride or a substituted alkenyl succinic anhydride or a mixture of these succinic anhydrides, and (b) 0.1-30 wt.% of phosphates of polyoxyethylene alkyl ether esters or phosphates of polyoxyethylene alkyl aryl ether esters or a mixture of these phosphate esters.

2. The composition according to claim 1 wherein the substituted alkyl succinic anhydride or substituted alkenyl succinic anhydride is additive reaction product of a straight chain inner olefin having 12 to 36 carbon atoms and maleic anhydride.

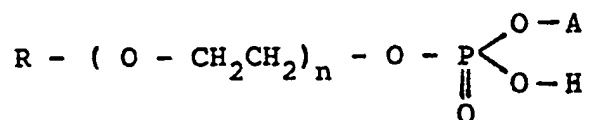
3. The composition according to any of claims 1 and 2 wherein the polyoxyethylene alkyl ether esters or phosphates of polyoxyethylene alkyl aryl ether ester are compounds represented by the following general formula:



where R is an alkyl or alkyl aryl group having 8 or more carbon atoms, A represents H, or

R' - (O - CH₂CH₂)_m - (R' is alkyl or alkyl aryl group, m is 1 or more integer number), and n is 1 or more integer number.

4. The composition according to claim 3 wherein compounds represented by the following general formula:



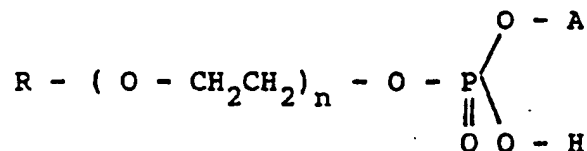
where the R of the above general formula is alkyl or alkyl aryl group having 10 - 20 carbon atoms and the value of n is 5 or more.

5. A process for using a composition for a sizing agent comprising dispersing the composition having (a) 70-99.9 wt.% of a substituted alkyl succinic anhydride or a substituted alkenyl succinic anhydride or a mixture of these succinic anhydrides, and (b) 0.1-30 wt.% of phosphates of polyoxyethylene alkyl ether esters or phosphates of polyoxyethylene alkyl aryl ether esters or a mixture of these phosphate esters in aqueous solution of aqueous soluble polymer compound, adding the produced aqueous dispersion to pulse slurry of paper making material, and making the paper.

6. The process according to claim 5 wherein the substituted alkyl succinic anhydride or a substituted alkenyl succinic anhydride employ an additive reaction product of a straight chain inner olefin having 12-36 carbon atoms and maleic anhydride.

7. The process according to any of claims 5 and 6 wherei

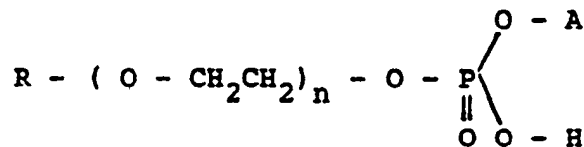
the polyoxyethylene alkyl ether esters or phosphates of polyoxyethylene alkyl aryl ether ester are compounds represented by the following general formula:



where R is an alkyl or alkyl aryl group having 8 or more carbon atoms, A represents H, or

$R' - (O - CH_2CH_2)_m -$ (R' is alkyl or alkyl aryl group, m is 1 or more integer number), and n is 1 or more integer number.

8. The process according to claim 7 wherein compounds represented by the following general formula:



where the R of the above general formula is alkyl or alkyl aryl group having 10 - 20 carbon atoms and the value of n is 5 or more.

INTERNATIONAL SEARCH REPORT

0151646

International Application No. PCT/JP84/00373

| | | |
|---|---|-------------------------------------|
| I. CLASSIFICATION OF SUBJECT MATTER (If several classification symbols apply, indicate all) ¹ | | |
| According to International Patent Classification (IPC) or to both National Classification and IPC | | |
| Int. Cl. ⁴ D21H3/08, 3/60 | | |
| II. FIELDS SEARCHED | | |
| Minimum Documentation Searched ⁵ | | |
| Classification System | Classification Symbols | |
| IPC | D21H3/02, 3/08, 3/60 | |
| Documentation Searched other than Minimum Documentation to the Extent that such Documents are Included in the Fields Searched ⁶ | | |
| | | |
| III. DOCUMENTS CONSIDERED TO BE RELEVANT ¹⁴ | | |
| Category ⁷ | Citation of Document, ¹⁶ with indication, where appropriate, of the relevant passages ¹⁷ | Relevant to Claim No. ¹⁸ |
| A | JP, A, 50-160505 (National Starch and Chemical Corp.), 25 December 1975 (25. 12. 75) & DE, A, 2,522,306 & FR, A, 2,272,221 | 1 - 8 |
| A | JP, A, 58-87398 (Idemitsu Kosan Co., Ltd.), 25 May 1983 (25. 05. 83) | 1 - 8 |
| A | JP, A, 58-45731 (Seiko Kagaku Kogyo Co., Ltd.), 17 March 1983 (17. 03. 83) | 1 - 8 |
| A | Nishi Ichiro and two others (authors) "Kaimen Kasseizai Binran" 5 July 1960 (05. 07. 60) Sangyo Tosho Kabushiki Kaisha See P. 20-21 | 1 - 8 |
| <p>¹⁵ Special categories of cited documents: ¹⁵</p> <p>"A" document defining the general state of the art which is not considered to be of particular relevance</p> <p>"E" earlier document but published on or after the international filing date</p> <p>"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)</p> <p>"O" document referring to an oral disclosure, use, exhibition or other means</p> <p>"P" document published prior to the international filing date but later than the priority date claimed</p> <p>"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention</p> <p>"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step</p> <p>"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art</p> <p>"G" document member of the same patent family</p> | | |
| IV. CERTIFICATION | | |
| Date of the Actual Completion of the International Search ¹ | Date of Mailing of this International Search Report ¹ | |
| October 5, 1984 (05. 10. 84) | October 15, 1984 (15. 10. 84) | |
| International Searching Authority ¹ | Signature of Authorized Officer ²⁰ | |
| Japanese Patent Office | | |